

THE GASIFICATION OF VARIOUS COALS IN MOLTEN SALTS

S. J. Yosim and K. M. Barclay

Energy Systems Group
Rockwell International
8900 De Soto Avenue
Canoga Park, California 91304

I. INTRODUCTION

The utilization of the U.S. coal reserves in a manner which does not add to the existing pollution problem is of utmost importance in the interest of conservation of more valuable natural resources in the national economy. Gasification of coal and generation of clean fuel gas offers one of the most promising approaches to the utilization of coal. It has been assigned a high priority in the U.S. Energy Development Program. Several of the coal gasification processes presently under development are now at the initial pilot plant operation stage. One of these processes is the Rockwell International Molten Salt Coal Gasification Process (Rockgas Process).^(1,2) In this process, the coal is gasified at a temperature of about 1800°F and at pressures up to 30 atm by reaction with air in a highly turbulent mixture of molten sodium carbonate containing sodium sulfide, ash, and unreacted carbonaceous material. The sulfur and ash of the coal are retained in the melt, a small stream of which is continuously circulated through a process system for regeneration of the sodium carbonate, removal of the ash, and recovery of elemental sulfur.

A molten salt coal gasification process development unit (PDU)^(1,2) capable of converting 1 ton of coal per hour into low-Btu fuel gas at pressures up to 20 atm is currently undergoing testing under contract to the Department of Energy. Preliminary to the PDU, a considerable amount of laboratory testing took place. These tests were conducted in a bench-scale, 6-in.-diameter gasifier in which coals of different rank were continuously gasified in the melt. The tests resulted in a better understanding of the gasification process. The purpose of this paper is to describe these laboratory tests and to discuss some of the chemistry taking place in the gasifier. Emphasis is placed on the effect of coal rank on the chemistry.

II. EXPERIMENTAL SECTION

A. COALS GASIFIED

The coals gasified were an anthracite, a medium-volatile bituminous coal, a high-volatile bituminous coal, and lignite. The coals are listed in order of decreasing rank. The first three coals were supplied by the Electric Power Development Corporation of Japan, and the lignite was supplied by Phillips Petroleum Company.

The proximate and ultimate analyses of the coals are listed in Table 1.

B. APPARATUS

A schematic of the bench-scale molten salt gasifier is shown in Figure 1. Approximately 12 lb of molten salt were contained in a 6-in.-ID and 36-in. high alumina tube placed in a Type 321 stainless steel retainer vessel. This stainless steel vessel, in turn, was contained in an 8-in.-ID four-heating-zone furnace. The four heating zones were each 8 in. in height, and the temperature of each zone was controlled by a silicon-controlled rectifier. Furnace and reactor temperatures were recorded by a 12-point Barber-Colman chart recorder.

TABLE 1
COMPOSITION OF COALS (WT %)

	Anthracite	Medium-Volatile Bituminous	High-Volatile Bituminous	Lignite
Proximate Analysis				
Moisture	2.78	2.26	0.85	32.46
Volatile Matter	4.92	30.36	38.71	28.70
Fixed Carbon	87.51	56.53	37.69	25.50
Ash	4.79	10.85	22.75	13.34
Ultimate Analysis				
Moisture	2.78	2.26	0.85	32.46
Carbon	85.27	71.85	62.26	35.34
Hydrogen	3.21	4.60	4.95	2.52
Nitrogen	0.81	0.78	0.82	0.96
Oxygen*	1.97	8.59	5.60	14.85
Sulfur	0.67	1.07	2.77	0.53
Ash	4.79	10.85	22.75	13.34

*By difference

The coal ground in a hand-turned burr mill was metered into the 1/2-in.-ID central tube of the injector by a screw feeder. Rotation of the screw feeder was provided by a 0- to 400-rpm Eberback Corporation Con-Torque stirrer motor. The coal was mixed in the injector with the air being used for gasification, and this coal-air mixture passed downward through the center tube of the injector and emerged into the 1-1/2-in.-ID alumina feed tube. This alumina feed tube was adjusted so that its tip was ~1/2 in. above the bottom of the 6-in.-diameter alumina reactor tube. Thus, the coal-air mixture was forced to pass downward through the feed tube, outward at its bottom end, and then upward through 6 in. of salt in the annulus between the 1-1/2-in. and the 6-in. alumina tubes.

III. RESULTS

A. PRODUCT GAS COMPOSITION FROM GASIFICATION WITH AIR

The test conditions for the gasification tests are listed in Table 2 which gives the melt temperature, the air and coal feed rates, the air/coal ratio, and the percent theoretical air. The last column shows the air feed as a percentage of the amount of air which is required to oxidize the coal completely to CO₂ and H₂O. The air/coal ratios and thus the percent theoretical air were chosen to give a good quality product gas from a heating value point of view. The steady-state composition and the higher heating value (HHV)* of the product gas obtained from the four coals are shown in Table 3. In each case, a good quality (>120 Btu/scf) low-Btu product gas was obtained. The product gas compositions were calculated on the basis of the carbon, hydrogen, and oxygen mass balance and assuming thermodynamic equilibrium for the water-gas shift reaction



To perform the mass balance, the coal analytical data shown in Table 1 were expressed in terms of an empirical formula, C_xH_yO_z. The results are shown in Table 3. The agreement between the observed and calculated values is, in general, quite good.

*The higher heating values include the heat of condensation of steam to liquid water.

TABLE 2
TEST CONDITIONS FOR GASIFICATION TESTS WITH COALS OF DIFFERENT RANK

Coal	Rank Number*	Melt Temperature (°F)	Air Feed Rate (scfm)	Coal Feed Rate (lb/h)	Air/Coal Ratio (scf/lb)	Percent Theoretical Air
Anthracite	1-2	1791	1.85	1.67	66.7	45.3
Medium-Volatile Bituminous	2-2	1773	1.85	1.97	56.4	44.2
High-Volatile Bituminous	2-5	1740	1.85	2.95	37.7	32.4
Lignite	4-1	1781	1.60	5.06	19.0	32.9

*The rank number shows the ASTM class number followed by the group number. In Class 1, 1-1 is higher than 1-2, etc.

TABLE 3
COMPARISON OF OBSERVED AND CALCULATED PRODUCT GAS COMPOSITION

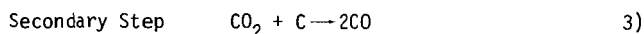
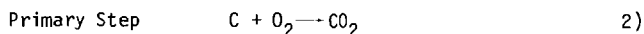
	Anthracite		Medium-Volatile Bituminous		High-Volatile Bituminous		Lignite	
	Observed	Calculated	Observed	Calculated	Observed	Calculated	Observed	Calculated
C ₀	29.9	28.9	28.7	26.2	31.4	33.4	24.3	22.2
H ₂	9.3	8.0	9.3	11.0	14.7	17.0	15.4	19.4
CH ₄	0.3	0.1*	0.0	0.1*	0.3	0.1*	1.7	0.9*
C ₂ H ₆	0.0	0.0*	0.0	0.0*	0.0	0.0*	0.2	0.0*
CO ₂	2.9	2.6	4.8	4.1	4.3	0.1	10.9	11.3
N ₂	58.6	60.4	57.3	58.6	49.5	49.5	44.6	46.2
Higher Heating Value (Btu/scf)	129.0	120.3	122.7	120.0	151.1	164.2	149.2	143.6

*Arbitrarily assumed values.

As expected, the heating value of the product gas increases as the percent theoretical air decreases. This can be seen in Table 3, where in the case of anthracite and medium volatile bituminous coals, a product gas resulted with an HHV of about 130 Btu/scf at about 45% theoretical air, and the high-volatile bituminous and lignite coals resulted in a product gas with an HHV of about 150 Btu/scf at about 32% theoretical air. However, these are practical lower limits as to the percent theoretical air which should be used. If the percent theoretical air is too low, there will not be sufficient oxygen to gasify all the carbon and the carbon content of the melt will continue to increase. This is most pronounced with high rank coals such as anthracite. In addition, if the percent theoretical air is too low, there will be insufficient heat released to the melt to sustain the operating temperature. This is most pronounced in the low rank coals such as lignite which contain a considerable amount of combined oxygen and moisture. Thus, there is a practical limit to the heating value that can be obtained for the product gas.

B. A MECHANISM OF COAL GASIFICATION

A certain amount of time was required for the heating value of the gas to exceed 100 Btu/scf; this time was different for coals of different rank. A plot of product gas heating value vs cumulative run time is shown for the four coals in Figure 2. It can be seen that the time for the product gas to reach a heating value >100 Btu/scf decreased with decreasing coal rank. In the case of the anthracite and the medium-volatile bituminous coal, the times were about 2 h and 1/2 h, respectively. The product gases from the lignite and the high-volatile bituminous coals both had initial heating values in excess of 100 Btu/scf with the lignite initially producing somewhat richer gas than the high-volatile bituminous coal. During the early stages of an experiment when the product gas heating value was increasing, it was found that the CO₂ concentration was initially very high and continued to decrease while the CO concentration was very low and continued to increase. It was also found that the carbon content of the melt increased with time. This effect is shown for the case of anthracite in Figure 3. This suggests that conversion of carbon to CO₂ is the primary step; reduction of CO₂ to CO by carbon in the melt is a secondary step.



The steady-state carbon contents of the melt are shown for the four coals in Table 4. The steady-state carbon content for lignite is only 0.3 wt% in contrast to 12% for anthracite. Thus, the lower the rank of the coal being gasified, the more reactive the carbon and the less free carbon in the bed necessary to promote CO production; hence, the time required to achieve steady state is shorter with lower rank coal.

TABLE 4
STEADY-STATE CARBON CONTENT OF MELT

Coal	Rank Number*	Steady-State Carbon Content of Melt (wt %)
Lignite	4-1	0.3
High-Volatile Bituminous Coal	2-5	2.4
Medium-Volatile Bituminous Coal	2-2	3.6
Anthracite	1-2	12.0

*The rank number shows the ASTM class number followed by the group number. In Class I, 1-1 is higher rank than 1-2, etc.

REFERENCES

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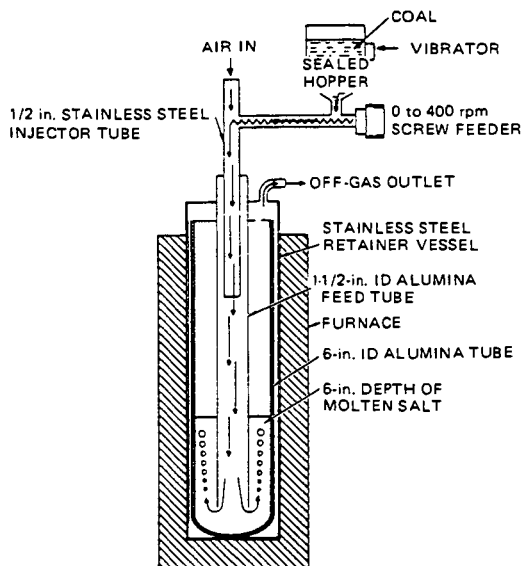


Figure 1. Bench-Scale Molten Salt Gasifier

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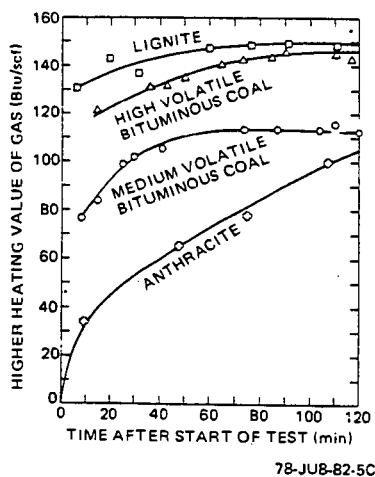


Figure 2. Change of Heating Value of Product Gas With Time for Different Coals

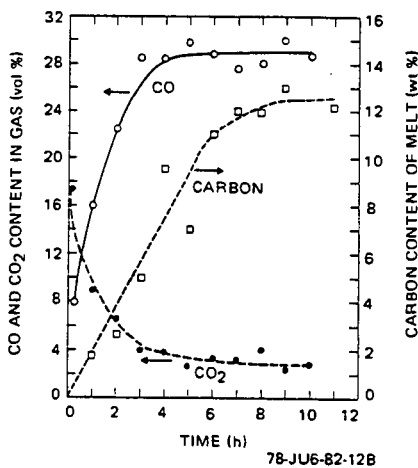


Figure 3. Effect of Carbon Content of Melt on CO and CO₂ Concentration in Gas